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Atomic and electronic contributions to Si(111)-(7×7) scanning-tunneling-microscopy images

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Bias-dependent scanning tunneling microscopy (STM), used in conjunction with atomic-charge superposition calculations, is shown to provide information on *both* atomic *and* electronic surface structure. Si(111)-(7×7) STM images measured with different bias voltages are compared with a number of recent structural models. Striking agreement is found with the stacking-fault–adatom model of Takayanagi. The unit-cell asymmetry found at negative sample bias is attributed to a stacking fault in one half of the unit cell, locally modifying the surface electronic structure.

Scanning-tunneling-microscopy (STM) images of the Si(111)-(7×7) surface were obtained two years ago by Binnig, Rohrer, Gerber, and Weibel and provided the first look at the overall atomic structure of this surface.[1] These first images have been interpreted to reflect the topology of the surface; several detailed models have been proposed based on these images and other experimental evidence.[1-6] However, recent theoretical[7] and experimental[8-10] work has shown that both electronic and atomic structure contribute to the STM images. In a recent STM paper Becker, Golovchenko, Hamann, and Swartzentruber clearly demonstrated that the empty surface states in the two halves of the Si(111)-(7×7) unit cell are different, which they argued might arise from the assumed presence of a stacking fault in one half of the unit cell.[9] However, a definite understanding of the observed differences could not be obtained, because the separation of structural and electronic contributions to STM images is a formidable problem and a major obstacle in quantitatively interpreting STM images. Theoretical calculations of STM images are possible once the electronic structure for the combined surface-tip geometry has been solved for a given surface geometry. To date, such calculations have not been performed for a realistic surface and a realistic tip. In addition, the dependence of the electronic structure on surface geometry makes this an impractical approach when dealing with uncertain or unknown geometries.

In this Rapid Communication we present a remarkably simple and general method to derive both geometric *and* electronic information from bias dependent STM images. Experimental STM images are compared with theoretical images calculated using the atomic-charge superposition method suggested by Tersoff and co-workers.[11] Since these calculated images do not contain surface-induced electronic states and only reflect the atomic geometry of the structure, one might expect the atomic charge superposition method to give a qualitative representation of STM images recorded at a bias voltage where surface states do not contribute strongly. We have found unanticipated *quantitative* agreement between calculated and measured images for Si(111)-(7×7) and Si(001)-(2×1) (Ref. 15) surfaces, which firmly establishes the value of such calculations. Deviations from this "geometric" image at other bias voltages can now be readily identified as due to electronic surface states and correlated with the geometric structure of the surface.

Here, we illustrate this approach for the Si(111)-(7×7) surface. We obtain excellent agreement with Takayanagi's model and demonstrate that other models proposed to explain the STM images are inconsistent with experimental results. In addition, we use this structure determination to spatially identify a modification in surface electronic structure associated with a stacking fault in the (7×7) structure.

In Fig. 1 we show representative STM images obtained for (a) negative and (b) positive bias voltages applied to the sample. These images are representative of a number of scans taken at bias voltages ranging from −1.2 to −2.7

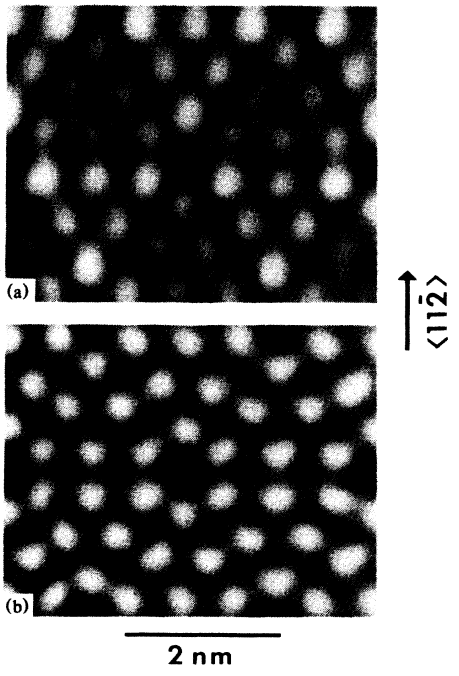

FIG. 1. STM images measured with (a) −2 V and (b) +2 V applied to the sample.

and +0.2 to +2.2 V. All experimental images shown here were obtained on *n*-type Si(111)-(7×7) samples using a microscope which has been described elsewhere.[12,13] The correction voltage applied to the piezoelectric crystal along the *z* axis (normal to the surface) to keep the tunneling current constant at 1 nA has been converted to the corresponding displacements of the tip in Å and plotted as a gray scale image. Here, high points are white, depressions black. The black-white range in this and subsequent images is 2 Å; the center of the gray scale corresponds to the average height in each case. This gray scale range was chosen to bring out all the relevant features in both these as well as the calculated images presented later. Experimental images have been digitally filtered with a low-pass noise filter on an off-line computer system.

In Fig. 1 we observe the orthorhombic (7×7) unit cell, containing twelve maxima inside the cell and a deep hole at the corners, in agreement with the results of Binnig *et al.*[1] We find that the strong left-right asymmetry of the unit cell [Fig. 1(a)] as originally observed by Binnig *et al.* occurs for negative bias on both *n*- and *p*- type Si(111)-(7×7) samples. For almost all positive bias voltages we did not find this asymmetry, although minor deviations from the result shown in Fig. 1(b) were observed in some narrow bias ranges.

In Fig. 2(a)–2(e) we present calculated STM images for a number of recent structural models of Si(111)-(7×7) using the atomic-charge superposition method suggested by Tersoff and co-workers.[11] In this approach an exponentially decaying, spherically symmetry charge density is assigned to each atom in the crystal. At any point the charge density can be calculated by summation of the charge densities of the atoms sufficiently close to contribute. As shown by Tersoff and Hamann, surfaces of constant charge density should be closely related to the corrugations measured in a STM experiment.[14] In addition to what we discuss next for the Si(111)-(7×7) surface, we find that calculations for the Si(001) dimer structure at the same charge density as used here (10^{-5} electrons/Å^3), show very good agreement with experimental results.[13,15] We have also compared results of the superposition method with charge densities calculated by self-consistent pseudopotential calculations for the Si(111)-(2×1) π-bonded chain model[16] and find that the two calculations give similar results. The charge density at which the corrugation was calculated was chosen to optimize agreement with the experimental results. The major change in going to higher (lower) charge density is an increase (decrease) in corrugation. For comparison we show an experimental image obtained with +2 V applied to the sample [Fig. 2(f)]. We also show line scans of measured (dotted lines) and calculated (solid lines) corrugations along the long (left) and short (right) diagonals connecting the corner holes of the (7×7) unit cell.

Figure 2(a) was calculated for the model by Binnig *et al.*, which consists of an array of twelve adatoms adsorbed on an unmodified substrate.[1] The adatoms give rise to the twelve maxima in the unit cell, which are well reproduced in the calculated image. However, the calculated corner holes are much shallower than in the experiment. Chadi proposed a variation of this model to explain the asymmetry of the unit cell observed by Binnig *et al.*, which he

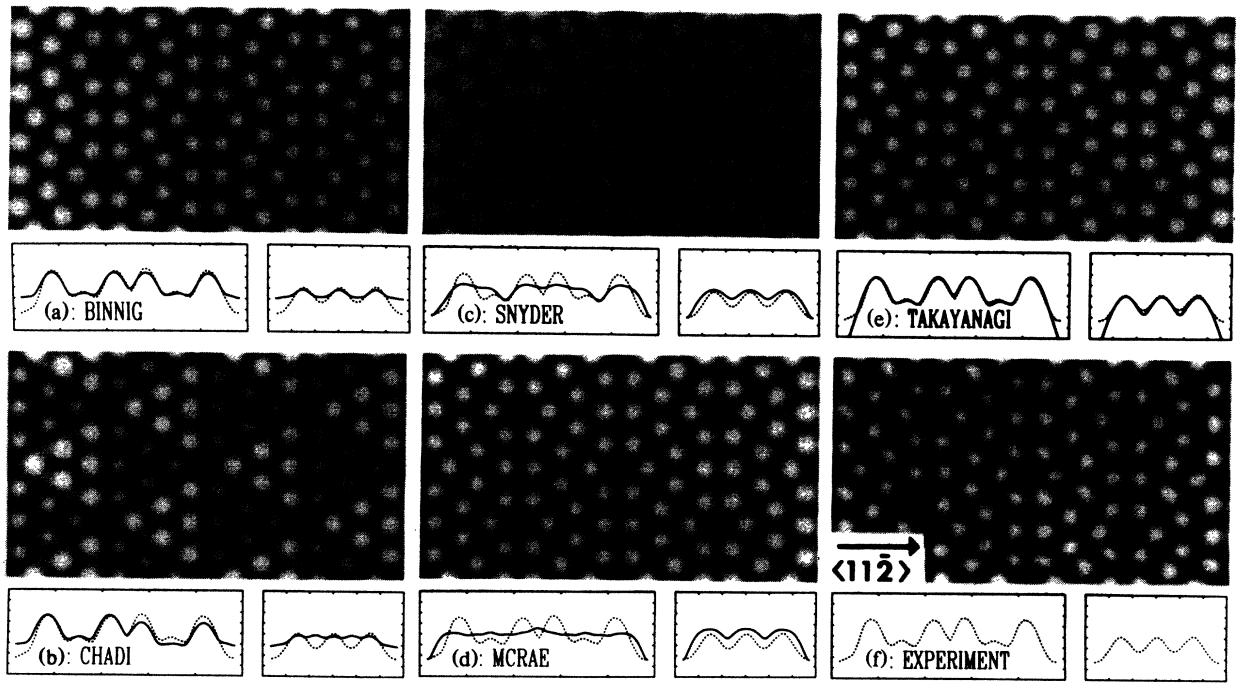

FIG. 2. (a)–(e) STM images calculated for (a) Binnig's model, (b) Chadi's model, (c) Snyder's model, (d) McRae's model, and (e) Takayanagi's model. (f) Measured STM image. The line scans run from corner hole to corner hole along the long (left) and short (right) diagonal of the (7×7) unit cell. The vertical range in these line scans is 4 Å. Solid lines are calculations, dashed lines represent the experimental results.

interpreted as an asymmetry in atomic corrugation.[3] In Fig. 2(b) this asymmetry is clearly observed, but also in this model the corner holes are too shallow and the corrugation along the short diagonal is very different from the experimental result. Snyder[4] and Aono[5] and co-workers proposed the pyramidal cluster model on the basis of cluster calculations, and low-energy ion scattering results, respectively. Instead of adatoms, clusters of four atoms are located on the surface in the same geometric arrangement as the adatoms in Binnig's model. Figure 2(c) shows the result for Snyder's model. Here, the overall symmetry of the minima seen in Figs. 1 and 2(f) is clearly absent, and because atoms in different clusters are very close to one another these clusters can hardly be resolved. In McRae's model [Fig. 2(d)] the outer two double layers contain two different stacking faults in the two halves of the unit cell.[6] The calculated image does not show 12 maxima in the unit cell, only structures along the lines connecting corner holes, corresponding to the dimers along the stacking faults. Finally, Fig. 2(e) is the result for Takayanagi's model.[2] It consists of a stacking fault in the outer double layer on half of the unit cell, which gives rise to the corner hole. On top of this substrate twelve adatoms are adsorbed. The agreement between the image calculated for this model and the experimental image shown in Fig. 2(f) is striking, particularly in view of the strong qualitative disagreement of the other models shown. The fact that the calculated corner hole is somewhat deeper than in the experiment is most likely a result of the tip being too large to completely fit into this deep and narrow minimum. Overall, the calculations agree with the experimental results to within 0.1 Å, which is remarkable in view of the approximations made in these calculations. We consider this exceptionally good agreement strong support for Takayanagi's model.

Recent studies of the geometric structure of the Si(111)-(7×7) surface by transmission electron diffraction (TED),[2,17] medium-energy ion scattering (MEIS),[18] and grazing angle x-ray diffraction[19] have also resulted in very strong support of Takayanagi's model. These studies have shown that many other models [like those presented in Figs. 2(a)–2(d)] cannot be reconciled with all experimental results, whereas quantitative agreement is found with Takayanagi's model.

As described above, the positive bias condition appears to reflect the atomic structure of the Si(111)-(7×7) surface. We recall that the charge superposition method does not take any electronic rehybridization effects into account, and calculated images are expected to reflect the atomic structure only. Although at first sight this seems a serious shortcoming of these calculations, the fact that only atomlike states are taken into account allows one to identify the contribution of surface-derived electronic states to the tunneling images. For example, the asymmetry of the unit cell seen for negative bias conditions in Fig. 1(a) does not appear to reflect the atomic geometry, but instead arises from an asymmetry in the electronic nature of the unit cell. In Takayanagi's model, one half of the unit cell contains a stacking fault in the outer double layer, just underneath the adatoms. In a recent pseudopotential calculation, Chou, Louie, and Cohen studied the electronic structure of stacking faults in Si.[20] They found that the stacking fault introduces a new state 0.1 eV above the top of the valence band at Γ and dispersing into the valence band away from Γ. Although this calculation was performed for a stacking fault in the bulk of a Si crystal, one would also expect a modification of the surface electronic structure associated with the presence of a surface stacking fault, giving rise to the observed asymmetry of the unit cell.

The experimental image shown in Fig. 1(a) shows additional features which we have not yet discussed. In the high half of the unit cell the three maxima next to the corner hole and the three maxima in the center are not equally high, the maxima at the corners being slightly higher. Although the adatoms in these positions all have an underyling stacking fault, they are geometrically inequivalent, which gives rise to differences in electronic structure as observed in the image.

In summary, we have shown that bias-dependent STM images can be used to extract *both* atomic *and* electronic information. By comparison of experimental results with atomic charge superposition calculations in which surface electronic contributions to the STM image are neglected, electronic and atomic contributions can be identified. Using this approach we have shown that Takayanagi's model agrees exceptionally well with STM, while other models are qualitatively inconsistent. The asymmetry of the images observed at negative bias voltages can be correlated with the presence of a stacking fault in one half of the unit cell. We believe that the approach we present here will be of general use to obtain a better understanding of the images obtained with scanning tunneling microscopy. In fact, the determination of a bias condition under which the STM image closely follows the atomic corrugations has been important in our recent current imaging tunneling spectroscopy experiments, in which energy resolved images of the Si(111)-(7×7) surface states were obtained with a lateral resolution of 3 Å.[21] In these experiments the variations of tunneling current with bias voltage in each point of the topographic image directly yield the surface electronic structure, separated from the geometric information. This separation is accomplished by having the tip follow the geometric corrugations as determined from a comparison of bias dependent STM images and atomic-charge superposition calculations as presented in this paper.

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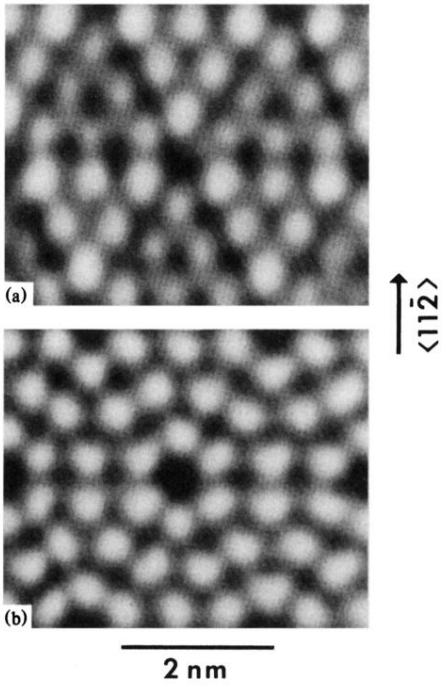
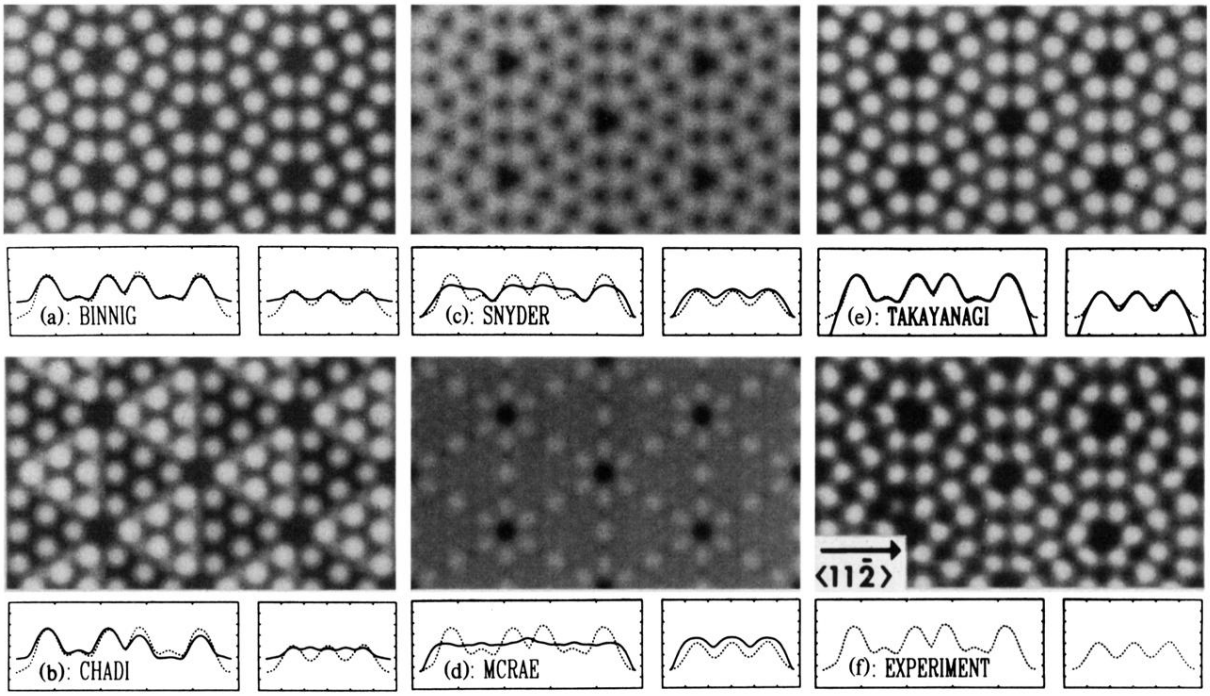


FIG. 1. STM images measured with (a) -2 V and (b) $+2$ V applied to the sample.



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